AD-A172 691 STUDY OF TRANSIENT PHONON DYNAMICS(U) INDIANA UNIV AT 1/1
BLOOMINGTON DEPT OF PHYSICS W E BRON 30 AUG 86
ARO-20446/17-PH DAAG29-83-K-0091

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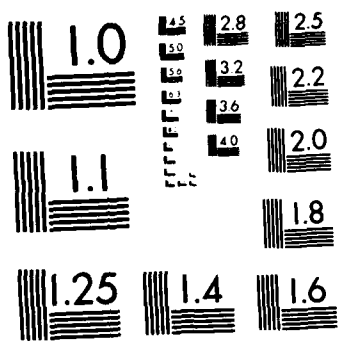

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2a. SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION/AVAILABILITY OF REPORT
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE	Approved for public release; distribution unlimited.
4. PERFORMING ORGANIZATION REPORT NUMBER(S)	5. MONITORING ORGANIZATION REPORT NUMBER(S) ARO 20446.17-PH
6a. NAME OF PERFORMING ORGANIZATION: Indiana University; 6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION: U. S. Army Research Office
6c. ADDRESS (City, State, and ZIP Code): Bloomington, IN 47405-6401	7b. ADDRESS (City, State, and ZIP Code): P. O. Box 12211 Research Triangle Park, NC 27709-2211
8a. NAME OF FUNDING/SPONSORING ORGANIZATION: U. S. Army Research Office; 8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER: DAAG29-83-K-0091
8c. ADDRESS (City, State, and ZIP Code): P. O. Box 12211 Research Triangle Park, NC 27709-2211	10. SOURCE OF FUNDING NUMBERS: PROGRAM ELEMENT NO. / PROJECT NO. / TASK NO. / WORK UNIT ACCESSION NO.

11. TITLE (Include Security Classification)
Study of Transient Phonon Dynamics

12. PERSONAL AUTHOR(S)
W. E. Bron

13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Year, Month, Day)	15. PAGE COUNT
Final	FROM 1 Jul 83 TO 30 Jun 86	Aug 30, 1986	15

16. SUPPLEMENTARY NOTATION
The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

17. COSATI CODES		
FIELD	GROUP	SUB-GROUP

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
Transient Phonon Dynamics, Lasers; Ionic Solids, Phonon Dynamics, Phonon Transport

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

The research program during the past three year grant period has been quite successful in devising new, and adapting existing, laser based techniques to the study of short and ultra-short duration laser induced transient dynamics. For the most part, work has concentrated on the dynamics of excited, nonequilibrium phonon distributions in semiconductors and in ionic solids. The excitation brings the solids well away from thermal equilibrium. It has been the author's contention that phonon dynamics and phonon transport can be investigated in greater detail by nonequilibrium techniques than by thermal equilibrium techniques as, e.g., thermal conductivity measurements. The results obtained so far offer an excellent opportunity for t expansion of existing methods and facilities to new areas of research.

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20. DISTRIBUTION/AVAILABILITY OF ABSTRACT ☐ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ DTIC USERS	21. ABSTRACT SECURITY CLASSIFICATION: Unclassified
22a. NAME OF RESPONSIBLE INDIVIDUAL	22b. TELEPHONE (Include Area Code) / 22c. OFFICE SYMBOL

ARO 20446.17-1

STUDY OF TRANSIENT PHONON DYNAMICS

FINAL REPORT

W. E. BRON

AUGUST 30, 1986

U. S. ARMY RESEARCH OFFICE

DAAG 29-83-0091

INDIANA UNIVERSITY

DEPARTMENT OF PHYSICS

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II. Review of Research Project and Accomplishments

A. General Remarks

The response of solids to laser light has been an active research area for a considerable period of time. The author's research program during the past three year grant period has been quite successful in devising new, and adapting existing, laser based techniques to the study of short and ultrashort duration laser induced transient dynamics. We have for the most part concentrated on the dynamics of excited, nonequilibrium phonon distributions in semiconductors and in ionic solids. The excitation brings the solids well away from thermal equilibrium. It has been the author's contention[1,2] that phonon dynamics and phonon transport can be investigated in greater detail by nonequilibrium techniques than by thermal equilibrium techniques as, e.g., thermal conductivity measurements. The results obtained so far offer an excellent opportunity for the expansion of existing methods and facilities to new areas of research.

The primary excitation produced in condensed matter by interaction with laser light spans the spectrum of relevance from basic physics to optical materials and devices. On the one hand, our investigation involves the dynamics of the coherent phonon state, the effect of electronic carriers on the coherent phonon state, on the nonlinear response of solids, and on the transport dynamics of phonons in solids. On the other hand, these investigations yield material parameters involved in the characterization of optical materials as to their ability to sustain short term nonequilibrium energy excursions and the

subsequent thermalization. The stability of, and the packing density of, optical devices will ultimately depend on these factors.

Experimentation was carried out in part at the picosecond and subpicosecond laser facility at the Max Planck Institute - Stuttgart, and at the facility currently being constructed under the author's supervision. The construction of the facility is funded (for the most part) through the DOD University Research Instrumentation Grant DAAG29-85-G-0106.

Studies of the transient dynamics of non-equilibrium phonons, which lay dormant only some 15 years ago, have advanced rapidly in the intervening time. A review of the accomplishments in the first decade may be found in reference 1. Over the past five years additional major advances have been achieved in the detection of stimulated phonon generation,[3-5] the possible detection of long lived transverse acoustic (TA) phonons,[6] the verification of the ν^{-5} scaling law for the lifetime of longitudinal acoustic (LA) phonons[7] (at least within the non-dispersive region), the discovery of new transport modes such as quasidiffusion,[8] the effect of surface and subsurface structures on phonon transport,[9] and frequency bunching of acoustic phonons.

During the more recent past, progress has also been made on the transient behavior of optical phonons and on electronic carriers and their mutual interaction.

The research activity described above, although limited to only a few laboratories, has examined new phenomena which are not

detectable by the older techniques of heat pulse transport or generation and detection with superconducting elements, or by measurements at thermal equilibrium, such as thermal conductivity. The source of this breakthrough has been the discovery of ever more sophisticated experimental techniques. The early methods which were limited to temporal resolution have since been replaced by techniques which permit simultaneous resolution in the spectral, spatial and temporal domains. Of highest importance has been the recent application of short and ultrashort laser pulses to studies of optical phonon dephasing and the interaction of these phonons with carriers and an electron-hole plasma. We have demonstrated that an electron-hole plasma at a conduction band minimum with concentration of $\lesssim 10^{16}$ pairs/cm^{-3} cannot, according to the above rule, interact directly with near-zone-center LO phonons with $q < 105$ cm^{-1}, although we demonstrate that it does interfere with coherent phonon excitation. It has also been demonstrated that in any event the direct LO phonon-plasma interaction is weak until the plasma concentration exceeds $\sim 5 \times 10^{17} cm^{-3}$.

B. General Experimentation

Most of the experimental techniques developed over the past 15 years do not involve the generation and/or detection of coherent phonon states. The generation and detection of coherent optical phonons has been demonstrated using coherent Raman excitation (CRE) and coherent anti-Stokes Raman (CARS) detection. In fact, this latter method is so versatile that it completes the search for a generally useful phonon spectrometer

operable over a range of ambient temperatures in each of the spatial, spectral, and temporal domains. Moreover, the technique can be used to detect dephasing effects. It does not require probe ions and is free of distorting surface effects. On the other hand, the method is limited, in first order, to excitations of optical modes near q-0. However, we have recently demonstrated an extension which includes acoustic phonons by coupling vibronic sideband spectroscopy to the CARS technique.

We turn to a review of some recent work on nonequilibrium transient dynamics. We consider here only those investigations, performed primarily by the author's research group.

C. Acoustic Phonons

A major step was the adaptation of the vibronic sideband phonon spectrometer (VSPS) to simultaneous spectral, spatial, and temporal resolution of nonequilibrium phonon transport. The full capability of the vibronic sideband phonon spectrometer (VSPS) has been demonstrated by Bron and Grill.[10] In these experiments laser induced luminescence from Eu^{2+} probe ions is used to determine the evolution of an injected phonon distribution propagating in a single crystal of SrF_2. The luminescence is excited in a focal column of a N_2 laser beam. The focal column can be placed anywhere inside the crystal, gaining, thereby, spatial resolution. Temporal resolution is obtained by delaying the luminescence signal by varying amounts relative to the time of the phonon injection. The early experimentation has recently been improved upon by Wilson, Lurie and Bron.[11] Various aspects of transient phonon dynamics are observed: Among

these are; (i) the "time of flight" spectrum follows that usually assigned to purely diffusive phonon transport, (ii) at the time of phonon injection immediately behind a thin film heater-substrate interface, the phonon distribution is almost a black body source but with marked deviations, (iii) the deviations from a Planck distribution disappear as the elapsed time after injection increases , (iv) in the interior of the sample and for $t < 2.25\ \mu s$ the mode temperature of the low frequency component of the phonon distribution is slightly higher than the high frequency component, with the latter component tending to form a local quasiequilibrium, and (v) as time progresses a more complete spatial quasiequilibrium begins to form. Moreover; (vi) components of the phonon distribution, which can interact anharmonically with each other during the experimental time span, reach a common mode temperature, (vii) only the lowest frequency component of the phonon distribution (which elastically scatters the least) can penetrate rapidly into the sample, but since low frequency phonons interact anharmonically only infrequently during the time span of the experiment their mode temperatures remain high compared to that of the quasiequilibrium, (viii) the high frequency component, because of strong elastic scattering, penetrates into the sample rather slowly at a rate determined by the diffusion constant of essentially the lowest frequency component of the quasiequilibrium. This form of phonon transport has been termed "quasidiffusive" tranport.

D. Laser Induced Acoustic Phonons

In our recent experimentation we have first used laser light

to generate monochromatic phonon distributions, and finally to both the generation and the detection of monochromatic phonon distributions. The advantages of laser light excitation over broad band excitation fixed at crystal surfaces was demonstrated through the use of FIR laser pulses to invert a three level electronic system in $Al_2O_3:V^{4+}$ to generate stimulated phonon emission of 740 GHz phonons.[3] Stimulated phonon emission excited by laser light has since also been observed using other electronic systems.[4,5] The same elecronic levels of V^{4+} in Al_2O_3 has also been used in conjunction with FIR laser light to generate initially monochromatic phonons at variable distances between the source and the detector. The resultant dynamics illustrated the features of quasidiffusive transport for the case of an initially monochromatic distribution of phonons.

In all of the above examples of studies of phonon dynamics either, or both, the source or detector depends on the presence of probe ions. Only a limited number of techniques have been developed so far which are applicable to "pure" (undoped) crystals.

E. Laser Induced Optical Phonons and Electronic Effects

In "pure systems" only optically (including Raman) active bulk modes near q=0 can be excited by laser light in first order. These modes have, however, lifetimes of the order of 10^{-12} seconds and, accordingly, require special experimental techniques if they are to be studied directly in the time domain. The rapid advance in the availability of synchronously pumped mode locked dye lasers with pulse widths of the order of picoseconds has made

it possible not just to excite and probe incoherent optical phonon distributions, but also to excite and probe coherent distributions through coherent Raman excitation (CRE) and detection by time resolved coherent anti-Stokes Raman scattering (TRCARS). This technique is not limited to any particular range of sample temperatures.

A part of our results and the experimental techniques was reported[12,13,14] in the form of short communications, in recognition of the rapid rate at which this field is expanding. More detailed manuscripts covering our results are in various stages of publication.

Among the results recently reported by the author and his collaborators are temperature dependence of the dephasing time ($T_2/2$) of coherent optical phonons in GaP and in ZnSe. The dephasing time of LO phonons in GaP at 5K was found to be 26 ps from which value it decreases to 6.7 ps at 300K. The measurements are made with a signal-to-noise ratio from 10^6 to 10^8. This highly advantageous signal-to-noise ratio is a consequence of coherent detection and of the high repetition rate (~ 76 MHz) of the mode locked lasers. As a result even weakly active processes may be detectable by this technique. The temperature dependence of $T_2/2$ in the regime $0 > T > 150$K can be fit to that expected from the time dependence of three phonon anharmonic processes. Above 120K the measured $T_2/2$ falls below the calculated values. It is expected that this deviation stems from dephasing through higher order phonon-phonon interactions. Contributions to the dephasing from boundary, impurty and carrier

scattering can be ruled out for the experimental conditions.

The early measurements of $T_2/2$ have since been extended in a number of ways. Among the salient features are that the dephasing time observed for coherent optical phonons is the same as that for incoherent ensembles. This result implies that the anharmonic operator acts on single optical phonon states even when the occupation probability is high. It remains to be seen what happens to this rule when the occupation probability of the acoustic phonons produced by the decay also are high. Similar measurements on coherent TO phonons show that the temperature dependence of the liftetime of these phonons is completely anomalous in GaP and to a lesser extent in ZnSe, whereas, it is normal in GaAs and for LO phonons in all these systems.

Further results include the determination of all the components of the complex third order nonlinear electronic susceptibility, $\chi^{(3)}$, for GaP[14] and ZnSe.

The basic assumption that three-phonon anharmonic processes dominate the decay of longitudinal phonons (LO) at low temperatures was next demonstrated. The topography of the dispersion relations of GaP suggest that decay proceeds solely to longitudinal acoustics (LA) such that $LO(\omega_{LO}, q\sim o) \longleftrightarrow LA(\omega_{LO})/2, q) + LA(\omega_{LO}/2, -q)$. Experimental verification of the above postulate was demonstrated by us by monitoring the presence of LO and LA phonons directly in the frequency domain through anti-Stokes VSPS measurements. The results clearly demonstrate that only $LO(\omega_{LO})$ and $LA(\omega_{LO}/2)$ phonons are present over a time span of 5 ns of LO phonon

generation. Theory predicts that the anti-Stokes sideband intensity, I_{AS}, should scale linearly with the power of either of the ℓ- or s-pump lasers. It was, however, observed that I_{AS} proceeds through a maximum (and returns to zero) as the power of either of the two lasers increases. This unexpected result is shown to arise from an interference which occurs, under the experimental conditions, between CRE and a nonlinear polarization resulting from the presence of an electron-hole plasma. The plasma is generated through the decay of "hot" carriers produced through a two step adsorption of the incident laser beams.

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13. Picosecond Laser Induced Transient Dynamics of Phonons in GaP and ZnSe, W. E. Bron, J. Krell and B. K. Rhee, submitted to Phys. Rev.

IV. List of Participating Scientific Personnel

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